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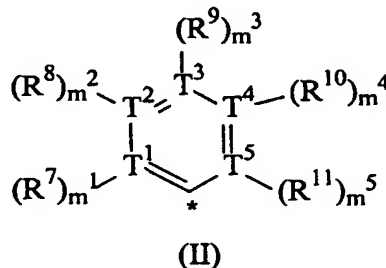
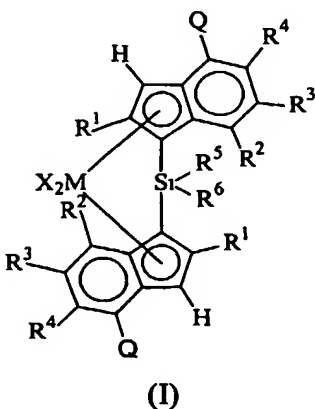
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ance Notes on Codes and Abbreviations" appearing at the begin-  
ning of each regular issue of the PCT Gazette.

(54) Title: PROCESS FOR THE COPOLYMERIZATION OF ETHYLENE



(57) Abstract: A process for producing a copolymer of ethylene containing from 0.1 to 99 % by mol of one or more derived units of alpha-olefins of formula  $\text{CH}_2=\text{CHA}$ , wherein A is a  $\text{C}_2\text{-C}_{20}$  alkyl radical, comprising contacting, under polymerization conditions, ethylene and one or more alpha-olefins in the presence of a catalyst system obtainable by contacting: a) a metallocene compound of formula (I): wherein M is zirconium, titanium or hafnium; X, same or different, is a hydrogen atom, a halogen atom, or an hydrocarbon group;  $\text{R}^1$ , is a hydrocarbon group;  $\text{R}^2$ ,  $\text{R}^3$ ,  $\text{R}^4$ ,  $\text{R}^5$  and  $\text{R}^6$ , are hydrogen atoms or hydrocarbon groups; Q is a radical of formula (II), being bonded to the indenyl.

at the position marked by the symbol \*; (II) wherein:  $\text{T}^1$ ,  $\text{T}^2$ ,  $\text{T}^3$ ,  $\text{T}^4$  and  $\text{T}^5$ , are carbon atoms (C) or nitrogen atoms (N);  $\text{m}^1$ ,  $\text{m}^2$ ,  $\text{m}^3$ ,  $\text{m}^4$  and  $\text{m}^5$  are 0 or 1;  $\text{R}^7$ ,  $\text{R}^8$ ,  $\text{R}^9$ ,  $\text{R}^{10}$  and  $\text{R}^{11}$  are hydrogen atoms or hydrocarbon groups; and b) an alumoxane or a compound capable of forming an alkyl metallocene cation.

# INTERNATIONAL SEARCH REPORT

Rec'd 29 MAR 2004

International Application No.

PCT/EP/02/40067

**A. CLASSIFICATION OF SUBJECT MATTER**  
EPC 7 C08F210/16 C08F4/642

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, CHEM ABS Data

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 01/48034 A (FISCHER DAVID ;WEISS HORST (DE); BINGEL CARSTEN (DE); FRAAIJE VOLK) 5 July 2001 (2001-07-05) example 9 page 87 - page 88; examples 90-97 claims 2,15-23	1-7,13, 14
X	WO 98/40416 A (GOERES MARKUS ;TARGOR GMBH (DE); BINGEL CARSTEN (DE); FRAAIJE VOLK) 17 September 1998 (1998-09-17) page 19 - page 23; examples 3-5; table 1 claims 2,10	1-14
X	WO 01/46274 A (BASELL POLYOLEFINE GMBH ;RAUSCHENBERGER VOLKER (DE); LILGE DIETER) 28 June 2001 (2001-06-28) claims 1,5	1-14
	- / - -	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

**\* Special categories of cited documents :**

"A" document defining the general state of the art which is not considered to be of particular relevance

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

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Authorized officer

Parry, J

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 03/10107

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 02/02575 A (EXXONMOBIL CHEM PATENTS INC) 10 January 2002 (2002-01-10) tables 3-5 claim 23	1-14
A	----- WO 00/44799 A (BREKNER MICHAEL JOACHIM ;TARGOR GMBH (DE); FRAAIJE VOLKER (DE); KR) 3 August 2000 (2000-08-03) page 38 - page 40; examples 3-6 -----	1-14

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/EP 03/10107

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1 - 14 (in part)

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R7 where applicable is C1 and R8-R11 are hydrogen.  
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2. claims: 1-14 (n part)

The subject matter of claims 1-14 (in part) where R7 where applicable is C2 and R8-R11 are hydrogen.  
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3. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R7 where applicable is C3 and R8-R11 are hydrogen.  
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4. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R7 where applicable is C4 and R8-R11 are hydrogen.  
---

5. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R7 where applicable is C5 and R8-R11 are hydrogen.  
---

6. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R7 where applicable is C6 or higher and R8-R11 are hydrogen.  
---

7. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R8 where applicable is C1 and R7,R9-R11 are hydrogen.  
---

8. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R8 where applicable is C2 and R7,R9-R11 are hydrogen.  
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9. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R8 where applicable is C3 and R7,R9-R11 are hydrogen.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

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10. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R8 where applicable is C4 and R7,R9-R11 are hydrogen.

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11. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R8 where applicable is C5 and R7,R9-R11 are hydrogen.

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12. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R8 where applicable is C6 or higher and R7,R9-R11 are hydrogen.

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13. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R9 where applicable is C1 and R7-R8,R10-R11 are hydrogen.

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14. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R9 where applicable is C2 and R7-R8,R10-R11 are hydrogen.

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15. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R9 where applicable is C3 and R7-R8,R10-R11 are hydrogen.

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16. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R9 where applicable is C4 and R7-R8,R10-R11 are hydrogen.

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17. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R9 where applicable is C5 and R7-R8,R10-R11 are hydrogen.

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18. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R9 where applicable is C6 or higher and R7-R8,R10-R11 are hydrogen.

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## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

## 19. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R10 where applicable is C1 and R7-R9, and R11 are hydrogen.

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## 20. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R10 where applicable is C2 and R7-R9, and R11 are hydrogen.

---

## 21. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R10 where applicable is C3 and R7-R9, and R11 are hydrogen.

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## 22. claims: 1-14 (in part)

the subject matter of claims 1-14 (in part) where R10 where applicable is C4 and R7-R9, and R11 are hydrogen.

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## 23. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R10 where applicable is C5 and R7-R9, and R11 are hydrogen.

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## 24. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R10 where applicable is C6 or higher and R7-R9, and R11 are hydrogen.

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## 25. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R11 where applicable is C1 and R7-R10 are hydrogen.

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## 26. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R11 where applicable is C2 and R7-R10 are hydrogen.

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## 27. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R11 where applicable is C3 and R7-R10 are hydrogen.

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## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

## 28. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R11 where applicable is C4 and R7-R10 are hydrogen.

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## 29. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R11 where applicable is C5 and R7-R10 are hydrogen.

---

## 30. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where R11 where applicable is C6 or higher and R7-R10 are hydrogen.

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## 31. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where any two of R7-R11 are not hydrogen and are not linked to each other.

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## 32. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where any two of R7-R11 are not hydrogen and form a ring.

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## 33. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where any three of R7-R11 are not hydrogen and are not linked to each other.

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## 34. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where any three of R7-R11 are not hydrogen and any two or more of them form a ring.

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## 35. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where any four of R7-R11 are not hydrogen and are not linked to each other.

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## 36. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where any four of R7-R11 are not hydrogen and any two or more of them form a ring.

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FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

37. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where all five of R7-R11 are not hydrogen and are not linked to each other.

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38. claims: 1-14 (in part)

The subject matter of claims 1-14 (in part) where all five of R7-R11 are not hydrogen and any two or more of them form a ring.

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

Intern Application No

PCT/EP 03/10107

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 0148034	A	05-07-2001	DE 19962905 A1	12-07-2001
			AU 2511001 A	09-07-2001
			BR 0016723 A	18-02-2003
			CA 2395552 A1	05-07-2001
			CN 1433435 T	30-07-2003
			WO 0148034 A2	05-07-2001
			EP 1250365 A2	23-10-2002
			JP 2003533550 T	11-11-2003
			US 2003149199 A1	07-08-2003
			ZA 200205003 A	23-06-2003
WO 9840416	A	17-09-1998	DE 19713546 A1	19-11-1998
			DE 19757563 A1	08-07-1999
			AT 249480 T	15-09-2003
			AU 6401098 A	29-09-1998
			AU 6727698 A	29-09-1998
			AU 7332398 A	29-09-1998
			BR 9808827 A	04-07-2000
			CA 2262396 A1	17-09-1998
			CA 2262493 A1	17-09-1998
			CN 1254328 T	24-05-2000
			DE 59809568 D1	16-10-2003
			WO 9840419 A1	17-09-1998
			WO 9840331 A1	17-09-1998
			WO 9840416 A1	17-09-1998
			EP 0968158 A1	05-01-2000
			EP 0942938 A1	22-09-1999
			EP 0931099 A1	28-07-1999
			JP 2001519779 T	23-10-2001
			NO 991112 A	07-05-1999
			NO 991113 A	07-05-1999
			TW 460495 B	21-10-2001
			US 2003009046 A1	09-01-2003
			US 6444606 B1	03-09-2002
			US 6492539 B1	10-12-2002
			ZA 9801912 A	06-09-1999
			ZA 9801913 A	06-09-1999
			ZA 9801915 A	06-09-1999
WO 0146274	A	28-06-2001	DE 19962130 A1	05-07-2001
			DE 10004660 A1	09-08-2001
			AU 1863201 A	03-07-2001
			BR 0016590 A	10-09-2002
			CA 2395218 A1	28-06-2001
			CN 1423664 T	11-06-2003
			WO 0146274 A1	28-06-2001
			EP 1110976 A1	27-06-2001
			EP 1252202 A1	30-10-2002
			JP 2001247729 A	11-09-2001
			JP 2004500452 T	08-01-2004
			US 2003130443 A1	10-07-2003
			US 2001004662 A1	21-06-2001
WO 0202575	A	10-01-2002	US 6380122 B1	30-04-2002
			US 6376413 B1	23-04-2002
			US 6380123 B1	30-04-2002
			US 6380121 B1	30-04-2002
			US 6380334 B1	30-04-2002

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 03/10107

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 0202575	A	US 6380331 B1	30-04-2002
		US 6380120 B1	30-04-2002
		US 6376407 B1	23-04-2002
		US 6414095 B1	02-07-2002
		US 6376409 B1	23-04-2002
		US 6376411 B1	23-04-2002
		US 6380330 B1	30-04-2002
		AT 260289 T	15-03-2004
		CN 1444593 T	24-09-2003
		CN 1444594 T	24-09-2003
		DE 60102156 D1	01-04-2004
		EP 1294733 A1	26-03-2003
		EP 1313746 A1	28-05-2003
		EP 1411058 A1	21-04-2004
		JP 2004502698 T	29-01-2004
		JP 2004502699 T	29-01-2004
		WO 0202575 A1	10-01-2002
		WO 0202576 A1	10-01-2002
		US 2003073568 A1	17-04-2003
		US 2002193242 A1	19-12-2002
		US 2002165084 A1	07-11-2002
		US 2002161143 A1	31-10-2002
		US 2003008767 A1	09-01-2003
		US 6376627 B1	23-04-2002
		US 6399723 B1	04-06-2002
		US 6376408 B1	23-04-2002
		US 6376410 B1	23-04-2002
		US 6376412 B1	23-04-2002
		US 6380124 B1	30-04-2002
WO 0044799	A	03-08-2000	
		DE 19903306 A1	03-08-2000
		AT 260945 T	15-03-2004
		BR 0004493 A	19-12-2000
		DE 59908742 D1	08-04-2004
		WO 0044799 A1	03-08-2000
		WO 0020466 A1	13-04-2000
		WO 0035973 A1	22-06-2000
		EP 1082363 A1	14-03-2001
		EP 1054914 A1	29-11-2000
		JP 2002532584 T	02-10-2002
		JP 2002535416 T	22-10-2002
		US 6486277 B1	26-11-2002
		US 6469114 B1	22-10-2002